Some methodological techniques for luminescence imaging using an IPD

C. J. McFee

Research Laboratory for Archaeology and the History of Art,
6 Keble Road,Oxford, UK.OX1 3QJ
(Present Address : Room 1016A, Building A2, Defence Evaluation and Research Agency, Farnborough, Hants GU14 0LX, UK.)

(Received 28 October 1997 ; in final form 6 May 1998)

Abstract: *An Imaging Photon Detector (IPD) can be used to obtain quantitative TL or IRSL images of the luminescence distribution in a mineral or pottery slice, or to measure the luminescence from individual mineral grains. Properties of the IPD itself, such as resolution and speed of response to the photon flux are important characteristics which will have important consequences for experimental design. However, the range of experiments that are possible in imaging studies are also dependent on the experimental system as a whole, such as the optics used, and the detector characteristics. In this paper the behaviour of such a system, using an IPD as the detector, is described, The consequences for the experimental methodologies adopted are also discussed, as are the range of measurements that are achievable from such a system.*

1. Introduction.

The lack of spatial discrimination of a photomultiplier (PM) tube denies the experimenter important information about the thermoluminescence (TL) or infra-red stimulated luminescence (IRSL) within a sample, whether it is a planchette containing many thousands of coarse-grains (90-180 μm) minerals, or a mineral or pottery slice. Only an integrated signal can be measured, and information on grain-to-grain or spatial variation in the luminescence is not available.

The obvious way of finding such information from single grains is by using a PM tube, picking and glowing out each grain individually (Miallier *et al.*, 1985, Grün *et al.*, 1989). However, the signal-to-noise ratio is often poor. To improve the signal to noise ratio larger grain sizes may be used. In addition, the photocathode can be physically or magnetically shielded from most of the thermal radiation from the heater plate, with only the photons originating from the area of the grain being measured (Wintle 1974, Sutton and Zimmerman, 1976). However, these measurements can be very time consuming and the need to illuminate the grains to be able to see them whilst picking can cause problems with IRSL measurements due to the highly light-sensitive nature of the IRSL traps (even if an image intensifier is used to view the grains whilst picking them). Recently, Bailiff *et al.* (pers. com.) have used a PM to detect the OSL signal stimulated by scanning a laser across a sample in a raster type scan pattern to build up an image of the OSL.

The problems associated with using a PM tube as the detector can be overcome using imaging detectors. Such techniques allow the luminescence distribution of mineral/pottery slices to be easily seen, and, if the luminescence distributions of individual grains are required, a large number of grains may be imaged in one measurement, saving a lot of time. However, it may be more difficult to obtain quantitative measurements using imaging techniques. In addition, the signal to noise ratio using imaging may not be as good as that obtained using a shielded PM tube or raster techniques.

Luminescence images may be found using photographic techniques, or by using a solid state imager. For example, Hashimoto *et al.* (1986, 1989), obtained TL colour images for a number of quartz samples and showed that it was possible to obtain quantitative TL information from these images. However, large (several kGy) doses had to be given to the quartz samples to maximise the TL counts. Huntley and Kirkey (1985) used an image intensifier to obtain the semi-quantitative distribution of the

natural TL from a quartz sample. An image intensifier was also used by Templer and Walton (1983) to study the TL from a number of zircon grains.

More recently, Duller *et al.* (1997) described a luminescence imaging system using a Charge Coupled Device (CCD) mounted on a Risø automated TL/OSL reader. This device enables quantitative luminescence images to be made at only a medium cost to the user. Integration effects limit the minimum time over which an image can be obtained (0.1s in this case). However, this is unlikely to be a major problem for the majority of measurements contemplated.

In this paper, the imaging techniques and methodologies adopted for luminescence imaging using an Imaging Photon Detector (IPD) are discussed. An IPD is a multichannel plate device which enables quantitative imaging of both TL (Smith *et al.* 1991, McFee and Tite 1998) and IRSL (McFee, forthcoming). The criteria adopted for selecting the optics which were eventually used with the IPD are described, followed by an examination of some of the potential problems which were found to be important in the optical arrangement used (and would lead to an incorrect measurement of the luminescence). Finally, two methodologies specifically adopted for imaging single grains are described and contrasted.

2. Maximising the resolution : the choice of optics.

An advantage of using a PM tube is that the tube is often placed directly above the sample to be measured. Thus, a large proportion of the available light is available to the photocathode. In contrast, in most imaging applications, optics will be required to focus the light onto the photocathode. This will reduce the overall aperture available for light collection. Therefore, great care needs to be taken when selecting suitable optics. For example, the optics initially used with the IPD were found to have a very poor resolution (Smith *et al.* 1991), with a 10mm aperture required to be placed in the optical path to remove any off-axis rays. However, this aperture heavily reduced the light throughput. Consequently, by considering the performance of the original optical system on the IPD, and by using a range of off the shelf lenses in a range of simple optical designs, a number of criteria were identified as being important in developing a new optical system for the IPD:

- the optics must be as aberration free as possible to allow the identification of individual coarse grains. Thus, grains would receive no additional light exposure above the minimum required for sample preparation and deposition;
- as large a depth of focus as possible is desirable as out of focus areas would lead to a significant loss of photons;
- a minimum working distance is necessary to accommodate a TL 'oven' (or an IRSL diode collar). With the oven used with the IPD, this distance was fixed at 15mm;
- a low magnification is usually desirable (<x10);
- as large a light gathering power and light throughput as possible is necessary to maximise the count rate.

The requirements above are difficult to satisfy in full. After considering a range of commercially available lenses it was decided that a reflecting objective, based on a Cassegrain system with a large primary mirror and a smaller secondary mirror, was the most appropriate lens. The advantages of this system were:

- zero chromatic aberration as there are no refracting optical elements (a problem with the previous lenses);
- a much longer working distance, for the same magnification, than with conventional refracting objectives;
- a large numerical aperture (0.5);
- wavelength range extends into the UV.

On the advice of an optical consultant, a suitable lens was purchased from Biorad, who manufacture microscope lenses for biological microscopes. The lens itself was an "off-the-shelf" lens, but had to be modified by the manufacturer to accommodate the working distance of 15mm.

Unfortunately, even after modification, the lens produced an image only 25mm above the housing of the lens, this was too short to project the image onto the photocathode of the IPD. Thus an additional 'relay' lens was required which produced a 1:1 image at the correct distance for the photocathode, the full optical set-up (i.e. reflecting objective and relay lens) is shown in figure 1. The secondary relay lenses were made of quartz with an effective transmission above 220nm, and were bought "off-the-shelf" from the Ealing Electro-optics catalogue. This final optical system was tested by visually imaging a series of test

bars in strong white light and the overall resolution of the system in white light was found to be better than 10μm with only small amounts of coma visible at the extremities of the field of view, no spherical aberration could be seen. The optical system was then tested by imaging a 180μm diameter pinhole at very low light levels (using a heavily filtered diode). These results are shown in table 1, together with the other specifications of the new and old optical systems.

	Original optics	New optics
Magnification of the optics	x 4	x 5.25
Best image size for the 120μm pinhole	600μm	180μm
Approximate depth of focus	n/a	300μm
Image field of view at the IPD Photo-cathode	25mm	19mm
Light Throughput (nominal values).	1 (with aperture)	14
Working distance of lens	n/a	25 mm
Effective object field of view	6.2mm	3.5mm

Table 1.
Measurements of the image size and depth of focus of the original optics, compared with the new values for the reflecting objective plus the secondary relay lens. The image sizes have been corrected for the magnification of the optics.

The resolution achieved with the new optics is more than sufficient to allow the imaging of either single grains or mineral/pottery slices, and the total light throughput of the optics has substantially improved. Although there has been a reduction in the image field of view, the grains can now be packed more closely together, due to the increased resolution. Consequently, the number of grains which can be imaged in a single measurement has not substantially declined.

The IPD is fitted with a bi-alkali photocathode, which is optimised towards the blue end of the spectrum. Although TL and IRSL measurements can be made with a range of filters, it was found that the use of blue sensitive filters (for example, a 7-59) limits the amount of light which falls onto the photocathode to the extent that, typically, only very bright grains can be seen. Consequently, the majority of measurements with the IPD were made using the broad-band BG39 filter. The transmission of the photocathode, combined with a BG39, is shown in figure 2.

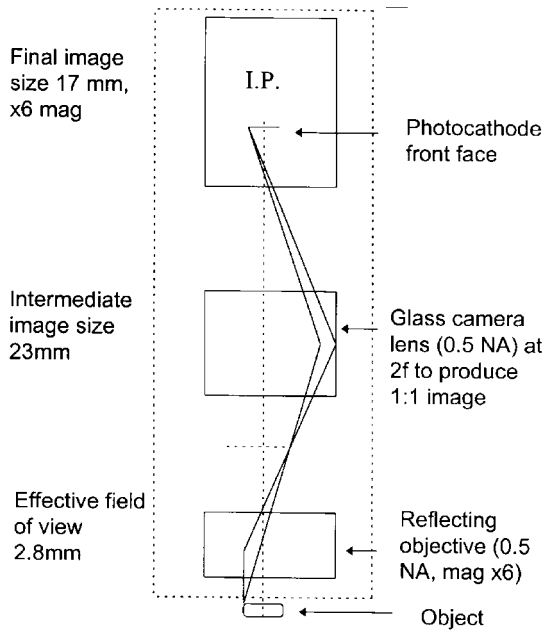

Figure 1.
The IPD optical setup

The light transmission of the IPD was compared with that of a "conventional" manual TL set (in which the PM tube is placed directly over the heating plate). The TL from a single coarse-grain of calcium fluoride was measured using both the "conventional" TL set, and the IPD). The IPD was found to be about 10 times less sensitive then this "conventional" set. The lower limit of detection for the IPD system varies, depending on grain size, sensitivity, etc. However, in general, most sediment samples were reasonably bright, and typically about half of the grains placed onto a planchette were visible when their natural TL was measured. In contrast, most pottery samples were very dim and only about 10 % at most were visible, even if the grains were given an appreciable radiation dose before glowing out. Similar results were found using IRSL. For typical sedimentary grains, about half of the grains which were placed onto the planchette had a measurable natural IRSL.

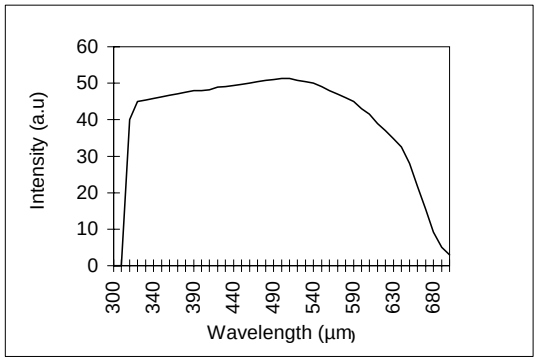

Figure 2.
The transmission of the quartz optics plus a BG39 optical filter.

3. Potential Problems from luminescence imaging.

Although a suitable optical system had been developed, a range of experimental factors needed to be addressed before an appropriate methodology could be defined. In particular, several major potential sources of error were identified as potentially leading to non-reproducible results when imaging mineral/pottery slices or single grains. These were:

- variation in transmission of the optics across the field of view;
- variation in photocathode sensitivity;
- variation in beta dose delivered to the slice/grains during an on-plate irradiation.

In addition, for single grains, changes in grain orientation between measurements may also lead to errors.

Each of these potential sources of error will be considered in turn.

3.1 Variation in transmission across the optics.

Figure 3 shows a flat field measurement across the IPD photocathode. It can be seen that the optics are not uniformly transmitting across the entire field of view. In addition, unless the IPD could be very accurately positioned above the sample, a small lateral displacement in position of the IPD or optics was found to lead to a large shift in image position on the photocathode. The causes of this shift in position are due to the current design of the IPD optical system. To allow samples to be placed onto the heater plate the entire IPD optical system (indicated by the dotted line in figure 1) can be raised and moved laterally to expose the heater plate. Although the IPD optical system was carefully constructed, it is not possible to re-place the IPD exactly in the same place with respect to the object on the heater plate. For example, a re-positioning of the IPD optical system of only 200µm displacement from the object will lead to an image displacement on the photocathode of over 1mm. Consequently, changes in the field of view of the optical system (see figure 3) could lead to substantial apparent differences in luminescence for otherwise identical measurements. However, a simple correction factor can be used to correct this observed luminescence, and a small calibration program was written to automatically assign a correction factor to any selected area, depending on its position within the field of view. Nevertheless, it is clear that, due to the rapid fall off in intensity near the edge of the field of view it is difficult to accurately apply a correction factor and large errors may be introduced. Thus, areas of interest near the periphery of the field of view were excluded from measurements to prevent erroneous measurements.

The sensitivity of the bialkali photocathode and the resistive anode will also vary with position and Smith *et al.* (1991) quote the variation from the IPD as ± 30%. However, for all measurements made here, any variation in the sensitivity of the IPD will have been included within the flat field correction made above.

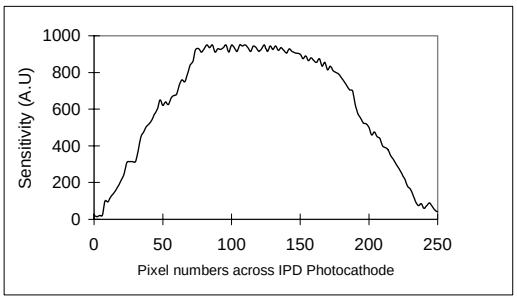

Figure 3.
A flat field measurement using the reflective objective and the secondary lens system. One pixel corresponds to approximately 0.1mm on the IPD photocathode.

3.2 Variation in beta dose across the sample during irradiation.

The beta dose rate delivered to a sample under laboratory irradiation will not be homogenous and there will be a diminution of the dose delivered at the extremities of a square planchette (10mm x 10mm). Aitken (1985, p123) quotes a difference of 10% in the dose between the centre and the extremes of a 10mm diameter stainless steel disc (for the seperation

of 2.5mm used here). However, the field of view of the new IPD optical system is less than 4mm. Thus, provided the sample is positioned correctly under the radiation source, it is likely that any differences in the beta dose rate across the field of interest will be insignificant compared to other possible sources of error discussed in this section. Consequently, they will be ignored.

3.3 Changes in grain orientation

The face which a grain presents to the photocathode will affect the total luminescence which can be measured. For example, Templer (1993) reported that the apparent TL sensitivity of a zircon grain could be altered by a factor of four, simply by turning a different face of the grain to the PM tube.

Possible errors due to changes in grain orientation were investigated by giving selected grains repeated cycles of a beta dose followed by a TL measurement, with each grain being viewed with a microscope between measurements, and then displaced with a needle to present a different face to the photocathode. In several cases the grains were physically removed altogether and then replaced. After every measurement, the TL from each grain was also corrected for any changes in position with respect to the field of view, as described in section 3.1 above. Several of the grains were specially selected because they had physical characteristics which it was thought would be most likely to show large variations in TL (for example, multi-faceted shapes or variable iron oxide staining on one face).

These measurements found that any variation in the TL due to the changes in grain orientation was no more than ±30% of the TL found in a single measurement.

In summary, several possible problems may occur in luminescence imaging. Variations in the beta dose in an irradiation applied to the area of interest were not considered important, whereas changes in grain orientation during a measurement could lead to errors in each measurement, but such errors are likely to be small (5-10%). The variations in transmission across the field of view, which could lead to large errors in the TL intensity measured in any single measurement, could be corrected for. However, this correction itself may also have a small error associated with it

Finally, as well as apparent differences in counts caused by a slight displacement of the image position on the photocathode, apparent changes in the TL could be found if small differences in the best focus postition occured, resulting in slightly different photon counts falling within an area of interest. Such differences would be particularly important when making measurements of the natural luminescence of single grains if inferences of, for example, the presence of any insufficiently bleached grains were made from these measurements .

The effect of each of these random variations is unlikely to be quantifiable. However, an empirical estimate of the total effect of such variations could be made by determining the range of TL intensities found from many grains after a repeated, identical measurement. Such an experiment is discussed below.

3.4 Measurement of the variation within a large number of grains

A large number of calcium fluoride grains were placed on a planchette. They were heated to 450 °C to zero the TL and were given a known laboratory beta dose. The grains were then glowed to 450 °C with a reheat to measure the blackbody radiation (glow 1). The grains were then given the same beta dose and glowed again (glow 2). The ratio of TL for glow 1/glow 2 were corrected for any positional differences for each grain within the field of view, as described in section 3.1. The ratios of glow1/glow2 are shown in figure 4 (a) and (b).

From this experiment, it can be seen that the TL intensities obtained from repeated, identical measurements may vary by up to a factor of four times (figure 4 (a)) if no correction for variation of grain position within the image field of view is applied. Applying the correction factor used in figure 3 reduces the largest difference in TL intensities obtained to 1.2 (figure 4 (b)).

The effect of applying a correction can be particularly seen for grains 180-200 where a large number of grains have a high ratio of glow1/glow2, presumably caused by a large movement of the image of the planchette within the field of view between measurements. This is greatly reduced after the correction has been applied.

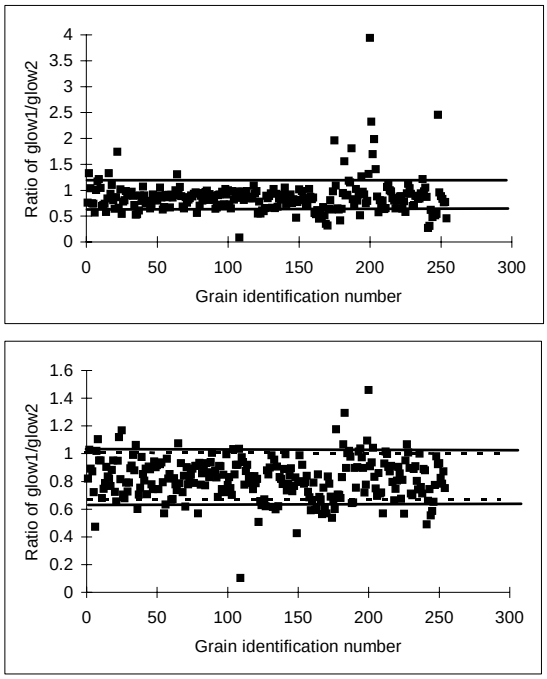

Figure 4.
(a, above) : The variation in the ratio of glow1/glow2 for 252 calcium fluoride grains; the TL measurements have not been corrected for changes in grain position due to planchette movement. The 1 sigma variation is shown
(b, below) : The same grains, but with a correction applied. The solid lines show the grains within ± 25% of the mean. The hatched lines show the 1 sigma variation. Note the changed scale.

The mean ratio for the grains without correction is 0.86 ±0.33 and with correction it is 0.82 ±0.15. The difference from the expected value of 1 is probably due to an increase in the TL sensitivity after the first TL measurement as the grains had no annealing treatment, before the zeroing of the TL by glowing to 450°C.

Figure 4 (b) also has plotted the 1σ standard deviation about the mean, and an arbitrarily chosen position which corresponds to ±25% around the mean value. When no correction is applied to the grains 187 out of 256 (73%) fall within ±25% of the mean, when a correction is applied it is 221 out of 256 (86%).

Therefore, the importance of correcting for differing planchette positions within the field of view is shown by figure 4. Applying a correction reduces the 1 sigma standard deviation by half, more importantly it corrects many potential outliers which would otherwise have large values of glow1/glow2.

However, despite this position correction the ratios of glow1/glow2 for all these grains do not share the same value. Although, after correction the number of grains falling in the ±25% band has increased, the variation in measurement values within this ±25% band appears quite similar. Most of the grains have been little affected by the correction for variation in planchette position, some not at all.

This experiment defined the minimum possible measurement reproducibility currently available from the IPD, which for the majority of measurements made was consequently taken as ±25%. This reproducibility is important because it points to the range of experiments that the IPD is very useful for, and conversely, those areas for which the IPD is not so suitable, these experiments are discussed further in section 5.

4. Methodologies adopted for imaging single grains

Although this paper has mainly concentrated on imaging single grains, the problems and solutions are just as valid for imaging mineral or pottery slices. However, for measuring single grains, one of two slightly different additional methodologies were adopted, the choice of methodology adopted depending on whether grains need to be physically identified after measurement, or if minimising the potential bleaching of any luminescence is more important. Both these methodologies will be discussed in turn.

method 1

Grains are hand picked with a small needle under subdued red light, using an image intensifier attached to a binocular microscope. The grains are then transferred onto a rhodium plated planchette which has small recesses drilled onto it to make a regular pattern (figure 5) to aid identification. The grains are not placed within the recesses but are grouped around the surface of the planchette surrounding each recess. It was found that placing a grain in a recess led to apparent differences in the TL intensities due to differences in reflectivity between the recess and the rest of the planchette. The TL image observed on the IPD is compared with the pattern seen when the planchette is examined with a microscope in reflected white light, after TL measurement.

Advantages of this method are:

- grains can be positively identified, allowing further investigations with, for example, a binocular microscope or an analytical SEM.

Disadvantages are:

- the considerable time needed to physically pick the grains and place them onto a rhodium planchette. As well as being very tedious, the grains will also be exposed to the subdued red light for an amount of time which could be sufficient to cause significant bleaching of samples, particularly if IRSL measurements are made;
- a limited number of grains may be imaged on each planchette.

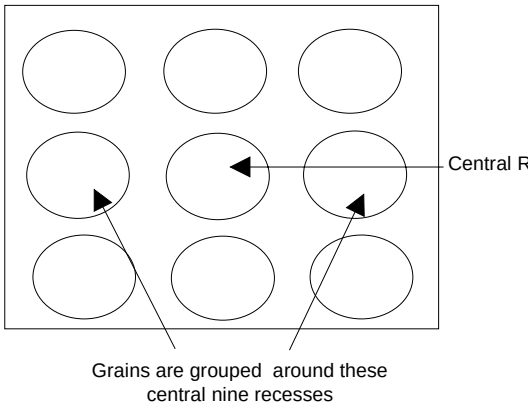

Figure 5.
The central portions of the rhodium plated planchette which are imaged by the IPD.

method 2

In this method the grains are not individually picked out but are prepared identically to the method adopted for preparing conventional stainless steel discs used for TL. A number of well-resolved grains may be selected from the TL image of the planchette.

Advantages of this method are:

- ease of preparation so that grains can be quickly prepared for measurement resulting in a saving of time and minimising possible bleaching;
- ability to measure more grains on a single planchette than in method 1.

Disadvantages of this method are:

- as the grains are randomly placed onto the planchettes some of them may be touching and consequently their images on the IPD cannot be completely resolved if there are insufficient counts to provide an adequate contrast between these grains. The data from these grains should be discarded;
- it is not possible to physically identify each grain after a TL measurement.

5. Conclusions

This paper has discussed some of the pitfalls that can occur in luminescence imaging, and the techniques adopted to try and overcome them. It is likely that some problems discussed in this paper are fundamental to luminescence imaging. For example, if single grains are to be measured, it is inevitable that the grains will have to be exposed to some light, unless the grains can be directly deposited on a disc and then resolved during the measurement.

However, it must be stressed that individual systems will have different problems associated with them. For example, an optical system with a larger field of view, or a system with less movement of the entire system between measurements, would not suffer the problems discussed in this paper to such an extent.

A careful assessment of the potential errors which could occur must be determined for individual systems, and this will determine to some extent the types of measurements that can be carried out with the system. For example, the IPD discussed in this paper was suitable for making measurements where very good reproducibility between measurements was not required. Thus, measurements of natural/second glow ratios to determine grains very bright in the natural TL were possible, as were diagnostic images of grains or pottery/mineral slices such as images of quartz using IRSL to investigate the possibility of feldspar inclusions within the grains (McFee, 1995).

However, when a good (within a few percent) measurement-to-measurement reproducibility is required, the IPD discussed in this paper is not so useful. For example, attempts to obtain single grain IRSL EDs proved only partially successful, using a weighted average it was possible to obtain an ED for a small number of grains that agreed with the ED found for the 'bulk' sample ED. However, the precision was such that it was not possible to distinguish between individual grain EDs (although a very high ED grain would still be clearly recognisable) (McFee, in press). Similarly, determination of fading rates for single grains was not possible with the system as it currently stands.

In summary, whilst luminescence imaging enables a wide range of measurements to be made, it is important to understand and characterise the behaviour of each individual system used. In addition, trade-offs within the choice of system used itself will also affect the type of measurements which are possible.

Acknowledgements

This work would not have been possible without the help of Adrian Allsop, Dave Seely and Martin Franks. Thanks to Lindsey Shepherd for help and comments on this paper.

References

Aitken, M.J. (1985) *Thermoluminescence dating*. Academic Press. London

Duller, G.A.T., Bøtter-Jensen, L., and Markey, B.G. (1997) A luminescence imaging system based on a charge coupled device (CCD) camera. *Radiation Measurements*, **27**, 91-99.

Grün, R., Packman, S.C., and Pye, K. (1989) Problems involved in TL-dating of Danish Cover Sands using K-feldspar. Long and Short Range Limits in Luminescence Dating, RLAHA Occasional Publication **9**.

Hashimoto, T., Koyanagi, A., Yokosaka, K., Hayashi, Y., and Sotobayashi, T. (1986) Thermoluminescence color images from quartzes of beach sands. *Geochemical Journal*. **20** , 111-118.

Hashimoto, T., Yokosaka, K., Habuki, H., and Hayashi, Y. (1989) Provenance search of dune sands using thermoluminescence colour images (TLCIs) from quartz grains. *Nucl. Tracks Radiat. Meas*., **16**, (1), 3-10.

Huntley, D.J., and Kirkey, J.J. (1985) The use of an image intensifier to study the TL intenstity variability of individual grains.*Ancient TL*. **3**(2) 1-4.

Miallier, D., Fain, J., and Sanzelle, S. (1985) Single grain quartz TL dating : an approach for complex materials. *Nucl. Tracks Radiat. Meas.*, **9**, 163-168.

McFee, C.J (1995) The Use of an Imaging Photon Detector for Luminescence Imaging Unpublished D.Phil thesis, University of Oxford.

McFee, C.J., (in press) The measurement of single grain IRSL EDs using an imaging photon detector. Accepted for publication in *Quaternary Geochronology*.

McFee, C.J., and Tite, M.S.. (1998) The detection of insufficiently bleached grains using an imaging photon detector. *Archaeometry* **40** (1), 153-168

Smith, B.W., Wheeler, G.C.W.S., Rhodes, E.J., and Spooner, N.A. (1991) Luminescence dating of zircon using an imaging photon detector. *Nucl. Tracks. Radiat. Meas*. **19**, 273-278.

Templer, R.H. (1993) Autoregenerative thermo-luminescence dating using zircon inclusions. *Archaeometry*, **35** (1), 117-136

Templer, R.H., and Walton, A.J. (1983) Image intensifier studies of TL in zircons. *PACT* **9**, 300-308.

Wintle, A.G. (1974) Factors determining the thermoluminescence of chronologically significant materials. Unpublished D.Phil. thesis, University of Oxford.

Sutton, S. R. and Zimmerman, D. W. (1976) Thermoluminescent dating using zircon grains from archaeological ceramics. *Archaeometry*, **18**, 125-134.

Reviewer :

H. Schwarcz